

## REMARKS

Claims 1-44 are pending in the present application. Claims 1, 2, 24, 30, and 43 have been amended. Amendments to claims 1, 2, 24, and 43 are supported in the specification, specifically p. 12, lines 6-7. The amendment to claim 30 is a deletion of an example provided in the claim thus no support from the specification is needed. No new matter is added by the claim amendments. Reexamination of the application and reconsideration of the rejections and objections are respectfully requested in view of the amendments and the following remarks, which follow the order set forth in the Office Action.

### *Information Disclosure Statement*

The Office Action indicates that the Andrews, Mayne, and Narducci references that were cited in an IDS by the Applicants were missing pages. Applicants are filing a supplemental IDS concurrently herewith submitting these references in their entirety.

### *Rejections under 35 U.S.C. § 112*

The Office Action rejected claims 2-3 and 30 under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter that applicant regards as the invention. More specifically, the Office Action asserted that the phrase "continuous or periodic automobile heat engine injector type" in claims 2-3 is allegedly indefinite. Applicants respectfully traverse.

Nonetheless, Applicants have amended claim 1 to recite a periodic injector system thus claim 2 has been amended to coincide with the amendment to claim 1. The recitation of a periodic injector system clearly defines the scope of claims 2 and 3. With regard to the recitation of an automobile heat engine injector type in the instant claims, Applicants assert that based on the disclosure provided in the specification, for example at page 12, lines 17-26, and the knowledge of one of ordinary skill in the art, one of ordinary skill would understand that which is being claimed in claims 2 and 3. As one of skill in the art would understand, all automobile heat engine injector systems primarily function in the same way; therefore, it is not necessary to limit the claims to a particular type of automobile heat engine injection system. Claims 2 and 3 specifically recite an automobile heat engine injector type injection system, thus the mentioning of a carbon black reactor is not applicable. A carbon black reactor is not an automobile heat injection system, as recited in claims 2 and 3, thus the

person of ordinary skill would not think that the injection system of claims 2 and 3 could possibly be a carbon black reactor.

Regarding claim 30, the Office Action asserted that the language “for example” is non-limiting. Applicants have amended claim 30 to remove the phrase “for example less than atmospheric pressure”. Applicants assert that the above claim amendments overcome the instant rejections and respectfully request reconsideration and withdrawal thereof.

### ***Rejections under 35 U.S.C. § 102***

#### **First § 102 rejection**

Claims 1-5, 8, 10-12, 16-23, 28-31, and 43-44 were rejected under 35 U.S.C. § 102(b) as being anticipated by Mayne, et al., *Pyrolytic production of aligned carbon nanotubes from homogenously dispersed benzene-based aerosols*, Chemical Physics Letters; 338: 101-107, (“Mayne”). Applicants respectfully traverse.

The instant claims relate to a method and a device for preparing carbon nanotubes or nitrogen-doped carbon nanotubes by pyrolysis. The method comprises using a periodic injection system to form liquid under pressure into finely divided liquid particles, conveying the finely divided particles by a carrier gas stream, and introducing the liquid into a reaction chamber where deposition and growth of the carbon nanotubes or nitrogen-doped carbon nanotubes takes place. The liquid contains at least one liquid hydrocarbon precursor of carbon or at least one liquid compound precursor of carbon and nitrogen consisting of carbon atoms, nitrogen atoms, and optionally hydrogen atoms and/or atoms of other chemical elements and optionally at least one metal compound precursor of a catalyst metal.

The periodic injection system provides benefits that were not available with previously disclosed methods. In particular, the periodic injection system enables the claimed method to be controllable, reliable, reproducible, and much more flexible than prior art methods, which employed a syringe, a sprayer, and an ultrasonic nebulizer. For example, the periodic injection system makes it possible to use any liquid based on any type of liquid hydrocarbon or liquid nitrogen compound, even liquids which have never been used or envisaged for making carbon nanotubes. Thus, it is possible to use solutions consisting of volatile carbon-containing liquids less toxic than benzene, although benzene may be used.

Additionally, the periodic injection system enables isolation between the “upstream” part of the method, e.g., the chamber for storing the liquid and the optional evaporator, and the heated reaction chamber. It enables the possibility of operating the reaction chamber with

a controlled pressure, which pressure may be less than atmospheric pressure. The flexibility of the periodic injection system enables the use of highly volatile liquids or less volatile and/or more viscous liquids. Additionally, the periodic injection system enables the use of different types of solutions with variable concentrations and even, surprisingly, the use of colloidal suspensions of metal particles where the size of the particles is generally less than 30 microns. The periodic injection system generates droplets with a well-defined volume and controls the liquid flow rate in a wide range, independently of the carrier gas flow rate. It enables a reproducible liquid flow rate, which is important in manufacturing scale production. See specification, pp. 8-9 and 12-18.

Mayne fails to disclose a fundamental element of the instant claims – the periodic injection system. It discloses a method of producing carbon nanotubes by pyrolyzing homogenously dispersed aerosols generated from benzene/ferrocene solutions at 800°C or 950°C using a compressed-gas-driven glass atomizer that works on the same principles as a commercial air-driven atomizer. *Mayne*, p. 102, Fig. 1. As disclosed in the instant specification, the operating principle of the atomizer is the same as that of a paint spraygun, which is fundamentally different than that of the claimed periodic injection system. See specification, p.6, lines 28-30 (English language translation). As explained in the instant specification, the use of spray generation systems, such as that used in *Mayne*, does not make it possible to generate large liquid delivery rates or to independently control the carrier gas delivery rate, the delivery rate of injected liquid, or the density of aerosol produced. Furthermore, these systems do not make it possible to use all types of solutions; in particular, solutions produced using volatile liquids. In contrast, the periodic injection system of the present claims overcomes the drawbacks that are present with the atomizer disclosed in *Mayne*.

Based on the foregoing, Applicants assert that *Mayne* fails to disclose each and every element recited in the instant claims. The periodic injection system avoids drawbacks and obstacles present in prior art methods as described in the specification on page 8-9 and 12-18 (especially pp. 16-18). As indicated above, in addition to simply being a different spray system from the periodic injection system of the instant claims, the atomizer disclosed in *Mayne* is unable to provide the above-described advantages. Accordingly, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

### **Second § 102 rejection**

Claims 1-5, 8, 10-12, 15-17, 20-23, 28-31, and 43-44 were rejected under 35 U.S.C. § 102(b) as being anticipated by Kamalakaran, et al., *Synthesis of thick and crystalline nanotube arrays by spray pyrolysis*, Applied Physics Letters 2000; 77(21): 3385-3387, ("Kamalakaran"). Applicants respectfully traverse.

Kamalakaran discloses a method of producing carbon nanotubes by spray pyrolysis of ferrocene/benzene solutions in an Ar atmosphere. *Kamalakaran*, p. 3385. The spray pyrolysis device comprises a pyrex nozzle attached at one end to a container for storing and releasing ferrocene/benzene solutions and attached at the other end to quartz tube. *Id.* The spray system of Kamalakaran does not make it possible to generate large liquid delivery rates or to independently control the carrier gas delivery rate, the delivery rate of injected liquid, or the density of aerosol produced. Furthermore, this system does not make it possible to use all types of solutions; in particular, solutions produced using volatile liquids. In contrast, the periodic injection system of the present claims overcomes the drawbacks that are present with the spray system disclosed in Kamalakaran.

Based on the foregoing, Applicants assert that Kamalakaran fails to disclose each and every element recited in the instant claims. Like Mayne, Kamalakaran fails to disclose a fundamental element of the instant claims – the periodic injection system. The periodic injection system avoids drawbacks and obstacles present in prior art methods as described in the specification on pages 8-9 and 12-18 (especially pp. 16-18). As indicated above, in addition to simply being a different spray system from the periodic injection system of the instant claims, the spray system disclosed in Kamalakaran is unable to provide the above-described advantages. Accordingly, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

### **Third § 102 rejection**

Claims 1-5, 9-12, 15-17, 20-22, 27-31, and 43-44 were rejected under 35 U.S.C. § 102(b) as being anticipated by Terrones, et al., *Novel nanoscale gas containers: encapsulation of N<sub>2</sub> in CN<sub>x</sub> nanotubes*, Chem. Commun. 2000: 2335-2336, ("Terrones"). Applicants respectfully traverse.

Terrones discloses a method of producing carbon nanotubes by spray pyrolysis of ferrocene/benzene solutions in an Ar atmosphere. *Terrones*, p. 2335. The spray pyrolysis device comprised a pyrex cone containing a nozzle, with the cone being attached at one end

to a container for storing and releasing ferrocene/benzene solutions and being attached at the other end to quartz tube. *Id.* As with the spray systems of Mayne and Kamalakaran, the spray system of Terrones does not make it possible to generate large liquid delivery rates or to independently control the carrier gas delivery rate, the delivery rate of injected liquid, or the density of aerosol produced. Furthermore, this system does not make it possible to use all types of solutions; in particular, solutions produced using volatile liquids. In contrast, the periodic injection system of the present claims overcomes the drawbacks that are present with the spray system disclosed in Terrones.

Based on the foregoing, Applicants assert that Terrones fails to disclose each and every element recited in the instant claims. As with Mayne and Kamalakaran, Terrones fails to disclose a fundamental element of the instant claims – the periodic injection system. The periodic injection system overcomes drawbacks and obstacles present in prior art methods as described in the specification on pages 8-9 and 12-18 (especially pp. 16-18). As indicated above, in addition to simply being a different spray system from the periodic injection system of the instant claims, the spray system disclosed in Terrones is unable to provide the above-described advantages. Accordingly, Applicants respectfully request reconsideration and withdrawal of the instant rejection.

### ***Rejections under 35 U.S.C. § 103***

The Office Action sets forth a series of § 103 rejections based on the primary references discussed above, Mayne, Kamalakaran, and Terrones, either alone or in combination with additional secondary references or official notice. Applicants respectfully traverse the § 103 rejections.

With the exception of the primary reference being different, the grounds of rejection are the same for all of the § 103 rejections. As can be seen from the arguments presented above, Applicants arguments with respect to the three primary references are the same, accordingly, in the interest of brevity and reduction of repetition, Applicants will respond to the § 103 rejections in masse rather than responding to the rejections based on each primary reference separately. For the sake of clarity, each of the § 103 rejections set forth in the Office Action is listed below. Applicants remarks in response to the § 103 rejections should be considered to be responsive to all of the § 103 rejections listed below.

- Claims 1-8, 10-12, 16-23, and 28-44 were rejected under 35 U.S.C. § 103 as being obvious over Mayne, either alone or in combination with each of the

following references, Zhu, et al., Direct Synthesis of Long Single-Walled Carbon Nanotube Strands, *Science* 2002; 296:284: 884-886 ("Zhu"); Ci et al., Preparation of carbon nanotubules by the floating catalyst method, *J. Mater. Sci. Ltrs.* 1999; 18: 797-799 ("Ci"); Cassell, et al., Large Scale CVD Synthesis of Single-Walled Carbon Nanotubes, *J. Phys. Chem. B* 1999; 103: 6484-6492 ("Cassell"); Anderson, U.S. Patent No. 5,697,342 ("Anderson"); Dai WO 00/63115 ("Dai"); and Li et al., Structure and growth of aligned carbon nanotube films by pyrolysis, *Chemical Physics Letters* 2000; 316: 349-355 ("Li CPL"); Li et al., Large-Scale Synthesis of Aligned Carbon Nanotubes, *Science* 1996; 274: 1701-1703 ("Li, Science"); Smiljanic, et al., Growth of carbon nanotubes on Ohmically heated carbon paper, *Chemical Physics Letters* 2001; 342: 503-509 ("Smiljanic"); Zheng, et al., *Chemical Vapor Deposition Growth of Well-Aligned Carbon Nanotube Patterns on Cubic Mesoporous Silica Films by Soft Lithography*, *Chem. Mater.* 2001; 13: 2240-2242 ("Zheng"); and official notice with regard to claims 32-42.

- Claims 1-8, 10-12, 16-23, and 28-44 were rejected under 35 U.S.C. § 103 as being obvious over Kamalakaran, either alone or in combination with each of the following references, Zhu, Ci, Cassell, Anderson, Dai, Li CPL, Li Science, Smiljanic, Zheng, and official notice with regard to claims 32-42.
- Claims 1-7, 9-17, 20-44 were rejected under 35 U.S.C. § 103 as being obvious over Terrones, either alone or in combination with each of the following references, Zhu, Ci, Cassell, Anderson, Dai, Li CPL, Li Science, Smiljanic, Zheng, and official notice with regard to claims 32-42.

As indicated in the remarks to the § 102 rejections, the injection systems disclosed in Mayne (atomizer), Kamalakaran (nozzle injection), and Terrones (nozzle injection) are completely different in form and functionality from the periodic injection system of the claimed invention. In addition to simply being different spray systems from the periodic injection system, the spray systems disclosed in the primary references are unable to provide the above-described advantages.

Furthermore, the injection systems of the primary references are all continuous injection systems. As discussed in the instant specification, the periodic injection system of the instant invention operates completely differently than a continuous injection system. The

term periodic generally means that the system carries out the injection discontinuously, opens periodically, and preferably operates at a fixed frequency. p. 12, lines 8-16 (English language translation). Thus, the disclosure of a continuous injection system would not make obvious to one of ordinary skill the use of a periodic injection system. Rather, such disclosure would tend to lead that person away from using a periodic injection system. Based on the foregoing, Applicants assert that none of the primary references make obvious the invention of the instant claims.

With regard to the cited secondary references, Applicants assert that none of the secondary references discloses a periodic injection system as recited in the claimed invention. As such, none of the secondary references overcomes the deficiency that is present in the primary references. Applicants address each of the secondary references more specifically below.

- Zhu discloses direct synthesis of long strands of single-walled carbon nanotubes by a catalytic chemical vapor deposition technique with a floating catalyst method in a vertical furnace. Zhu fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Ci discloses production of carbon tubules with diameters between 10-100 nm by the floating catalyst method using benzene and hexane as the hydrocarbon liquid. Ci fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Cassell discloses synthesis of single-walled carbon nanotubes by the chemical vapor deposition of methane using optimized catalysts. Cassell fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Anderson discloses a hydraulically-actuated fuel injector with direct control needle valve, especially for a direct-injection diesel-cycle internal combustion engine. Col. 3, lines 3-6. Applicants assert that the technical field of Anderson is far remote from that of the instant invention and thus non-analogous. It should not have been cited against the present application because it is in a field that is different from that of Applicants' endeavor and it

would not have commended itself to the attention of one of ordinary skill in the art in considering the claimed invention as a whole. MPEP § 2141.01(a). Further, there is no disclosure in Anderson of other uses or applications for the injector thereof. There is no indication in Anderson that would have led one of ordinary skill in the art to use the injector thereof for other purposes in other technical fields, let alone the very specific technical field of carbon nanotubes and their production. As such, Applicants assert that the conclusion of obviousness based on the combination of Anderson with any of the primary references is based on improper hindsight reasoning, which is forbidden. MPEP §2145 (X)(A). The Federal Circuit has cautioned that the determination of obviousness cannot be made by picking and choosing elements from available references using the "blueprint drawn by the inventor" in the application specification. *Interconnect Planning Corp. v. Feil*, 774 F.2d 1132 (Fed. Cir. 1985). Rather, the determination must be made based on the state of the art at the time of the invention. The Court also cautioned "[I]t is impermissible to use the claimed invention as an instruction manual or 'template' to piece together the teachings of the prior art so that the claimed invention is rendered obvious. ... This court has previously stated that '[o]ne cannot use hindsight reconstruction to pick and choose among isolated disclosures in the prior art to deprecate the claimed invention.'" *In re Fritch*, 972 F.2d 1260 (Fed. Cir. 1992). Because impermissible hindsight reasoning has been used to arrive at this conclusion of obviousness, Applicants submit that the claimed invention is not obvious over any of the primary references in combination with Anderson.

- Dai discloses pyrolyzing hydrocarbon gases in the presence of iron phthalocyanine to produce aligned nanotubes. Abstract and Fig. 1. As discussed in the instant specification, the synthesis technique of Dai involves vaporization of solid iron phthalocyanine stored in a boat placed in a furnace. This method prevents reproducible and stable vapor delivery rates from being obtained. Even in the form of a vapor, it is likewise difficult to convey these products continuously with a constant delivery rate, and this is a problem for application to continuous or semi -continuous production. The claimed method overcomes the problems present in methods such as those disclosed in



Dai by using a novel method of carbon nanotube production. Furthermore, Dai fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.

- Li CPL discloses a method for preparation of carbon nanotubes by pyrolysis of iron(II) phthalocyanine, under Ar/H<sub>2</sub> atmosphere at a predetermined temperature using an appropriate substrate in a flow reactor consisting of a quartz glass tube and a dual furnace fitted with independent temperature controllers. This reference is directed to reporting the detailed growth mechanism of the aligned nanotubes. Li CPL fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Li Science discloses a method for producing ordered, isolated, long carbon nanotubes based on chemical vapor deposition using mesoporous silica containing iron nanoparticles embedded in the pores as the substrate. Li Science fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Smiljanic discloses a method for producing carbon nanotubes using carbon vapor deposition wherein a carbon paper substrate loaded with Ni-Co or Fe catalyst is disposed perpendicular to an outer quartz tube of a reactor. Smiljanic fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Zheng discloses production of carbon nanotubes using carbon vapor deposition. The disclosed method uses a micromolding in capillaries technique combined with three dimensional cubic mesoporous silica films containing iron nanoparticles as catalysts to fabricate carbon nanotubes. Zheng fails to disclose any injection system whatsoever; therefore, it cannot possibly be effective in overcoming the deficiencies of the primary references in this regard.
- Official Notice – The Examiner took official notice that growing nanotubes on a variety of substrates with a catalyst is old and known. Even if such official

notice is correct, which Applicants are not agreeing with, such official notice still fails to overcome the deficiency in the primary references with regard to a periodic injection system. Thus, the instant claims are not obvious in view of any of the primary references in combination with the Examiner's official notice.

Based on the foregoing, Applicants assert that the claimed invention is not obvious over any of the cited references, either alone or in combination with one another. Accordingly, Applicants respectfully request reconsideration and withdrawal of the instant rejections.

For the foregoing reasons, claims 1-44 are considered allowable. A Notice to this effect is respectfully requested. If any questions remain, the Examiner is invited to contact the undersigned at the number given below.

Respectfully submitted,

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